Probing Thin Film Thermophysical Properties using the Femtosecond Transient ThermoReflectance Technique

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14. ABSTRACT
? Discuss Femtosecond Transient ThermoReflectance (FTTR) technique as a method for measuring the thermophysical properties of thin film materials. ? Demonstrate the measurement of the thermal diffusivity, electron-phonon coupling factor, and thermal boundary resistance of thin metallic films using the FTTR technique. ? Discuss the importance of considering the nonlinear relationship between reflectance and temperature. ? Present experimental results for Femtosecond Transient ThermoTransmittance (FTTT) studies performed on amorphous silicon solar cells.

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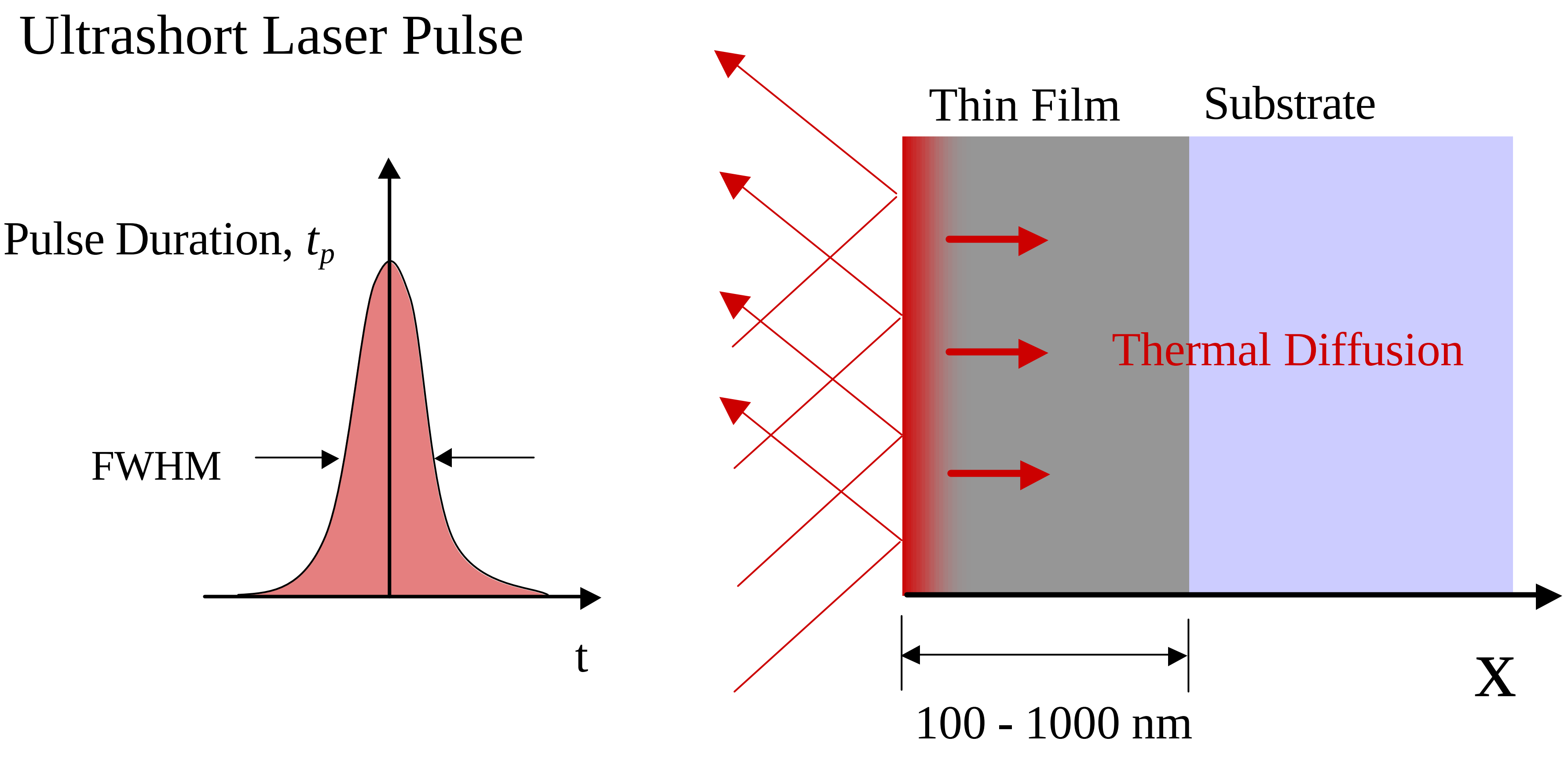
Objectives

- Discuss Femtosecond Transient ThermoReflectance (FTTR) technique as a method for measuring the thermophysical properties of thin film materials.
- Demonstrate the measurement of the thermal diffusivity, electron-phonon coupling factor, and thermal boundary resistance of thin metallic films using the FTTR technique.
- Discuss the importance of considering the nonlinear relationship between reflectance and temperature.
- Present experimental results for Femtosecond Transient ThermoTransmittance (FTTT) studies performed on amorphous silicon solar cells.

Transient Thermal Property Measurement



Nonequilibrium Energy Deposition Process

Free Electrons Absorb Radiant Energy

↓

Electrons Transfer Energy to the Lattice

Thermal Diffusion by Hot Electrons

Electron-Phonon Coupling Factor

↓

Thermal Equilibrium (~1 ps)

↓

Thermal Diffusion within Thin Film

Thermal Diffusivity

↓

Thermal Conductance across the Film/Substrate Interface

Thermal Boundary Resistance

↓

Parabolic Two Step (PTS) Model

Electron System:

$$C_e(T_e)\frac{\P T_e}{\P t}=\frac{\P}{\P x}\left(K_e(T_e)\frac{\P T_e}{\P x}\right)-G\left[T_e-T_l\right]+S(x,t)$$

Lattice System:

$$C_l\frac{\P T_l}{\P t}=G\left[T_e-T_l\right]$$

Electron Heat Capacity:

$$C_e\left(T_e\right)=gT_e$$

Electron Thermal Conductivity:

$$K_e\left(T_e,T_l\right)=K_{eq}\frac{T_e}{T_l}$$

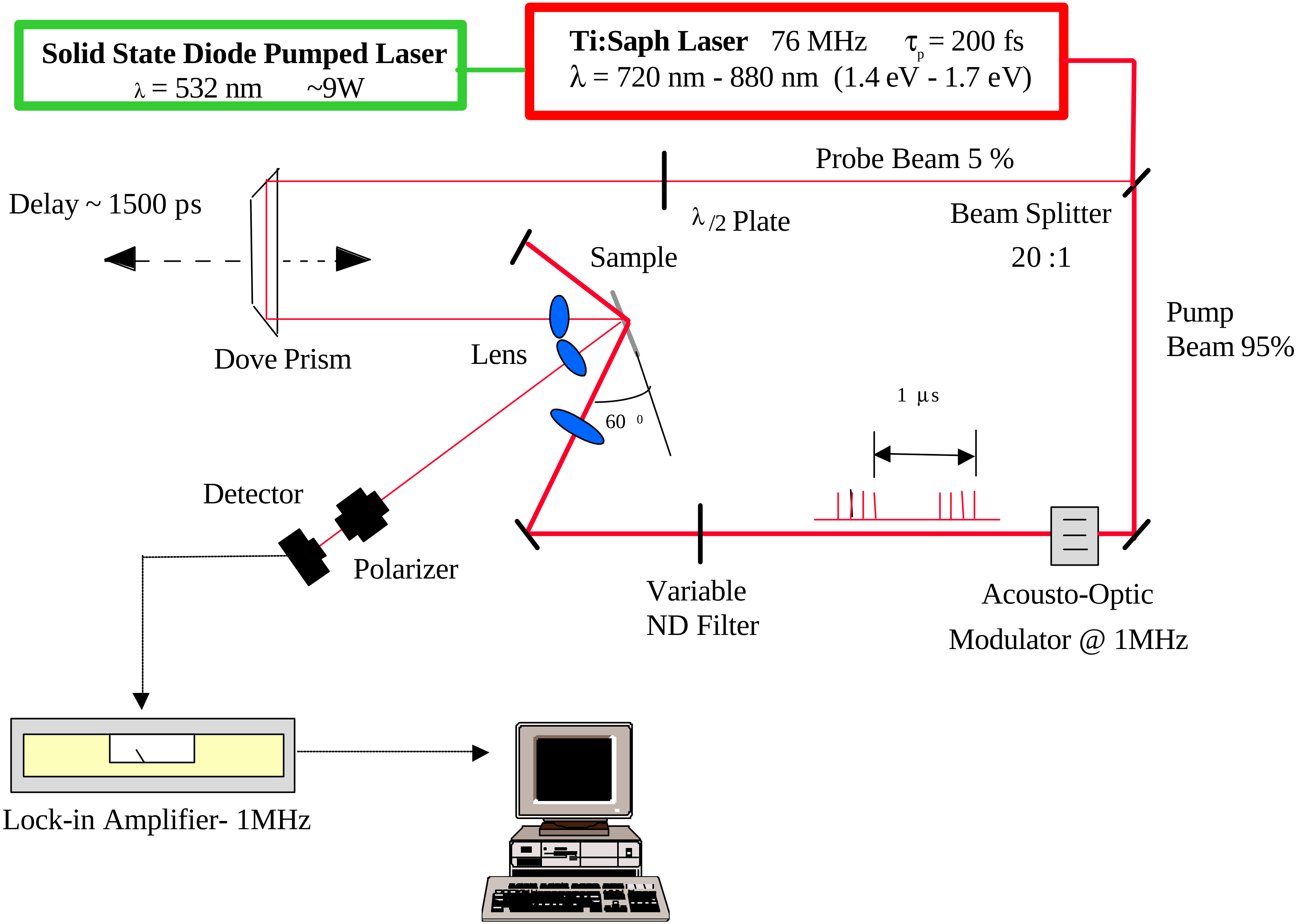
***G* = Electron Phonon Coupling Factor**

C_l = Lattice Heat Capacity

g = Electron Heat Capacity Constant

$S(x,t)$ = Laser Source Term

Transient ThermoReflectance Technique



Reflectivity as a Function of Temperature and Strain

Laser Absorption

Nonequilibrium Temperature Rise

Electron-Phonon Coupling

Thermal Diffusion

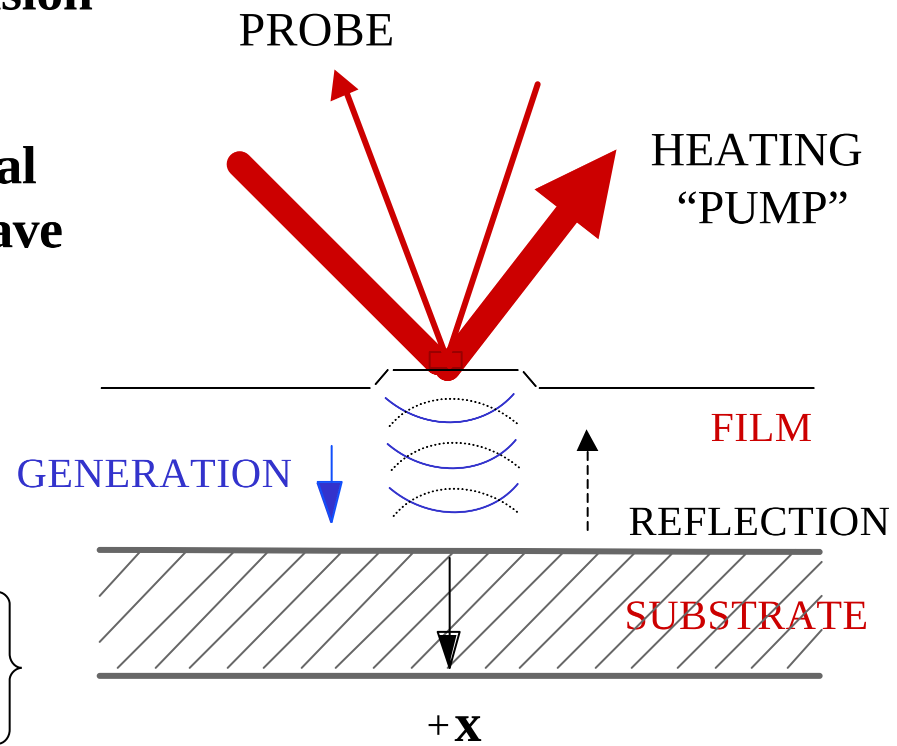
$D\boldsymbol{R}(\boldsymbol{T_e},\boldsymbol{T_l})$

$$\left\{ \frac{DR}{R} = aDT_e + bDT_l \right\}$$

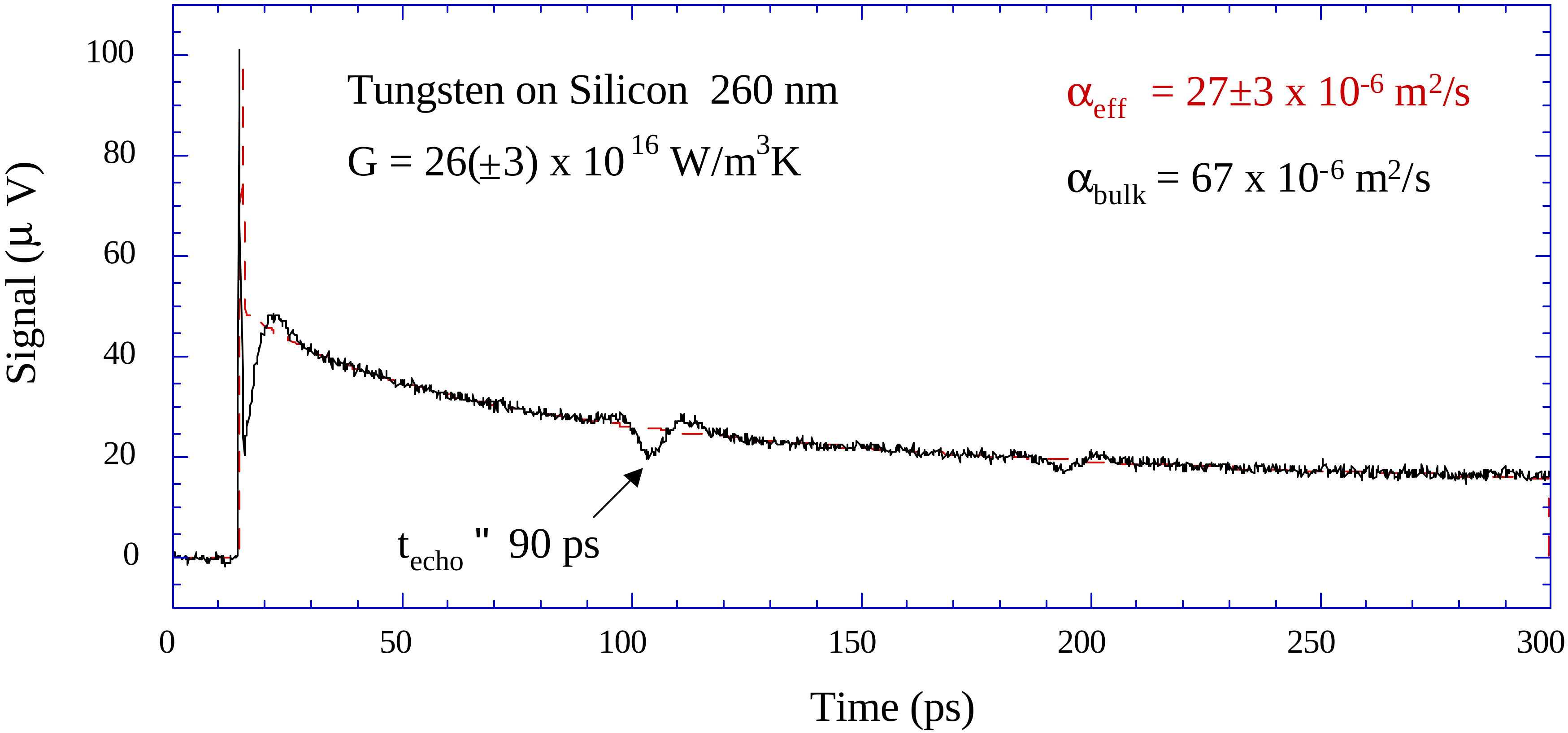
Thermal Expansion

Compressional "Acoustic" Wave

$D\boldsymbol{R}(h)$

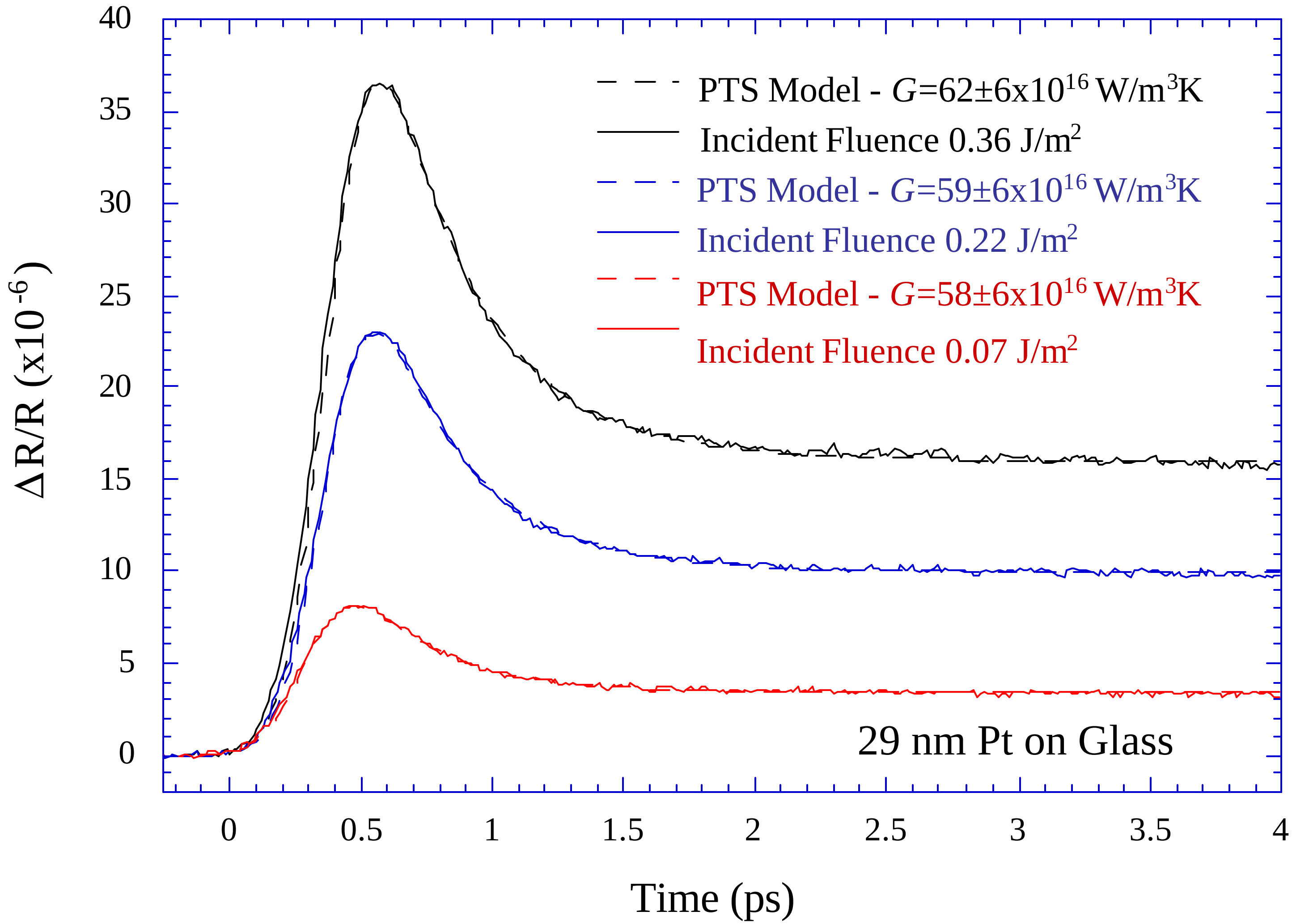


Thermal Diffusivity (W)



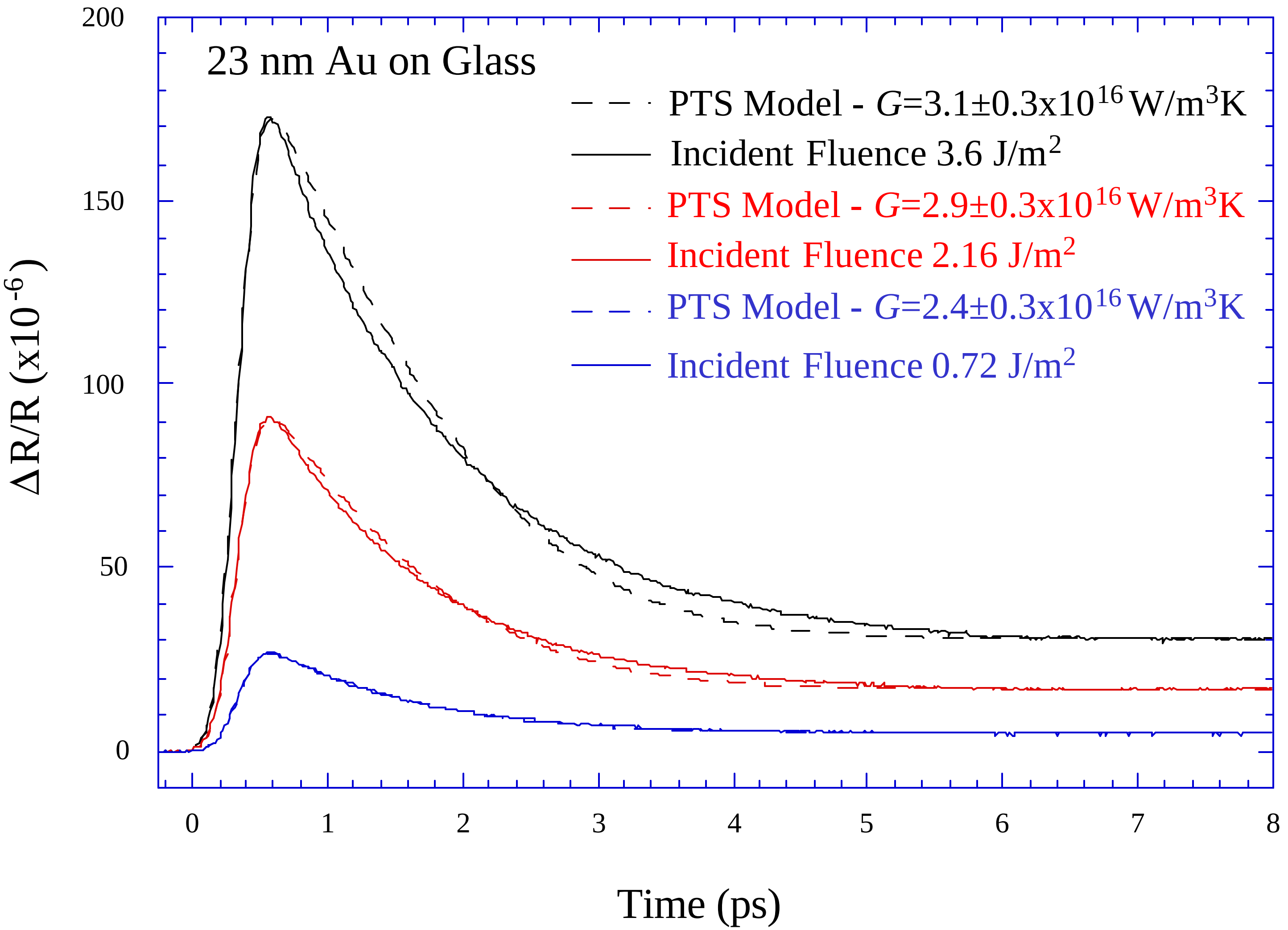
- The large initial response represents the initial nonequilibrium electron temperature.
- The ultrasonic echoes allow for the measurement of the sound velocity and/or elastic constants assuming the film thickness is known.
- The diffusion of thermal energy allows for determination of the thermal diffusivity.

Electron Phonon Coupling Factor (Pt)



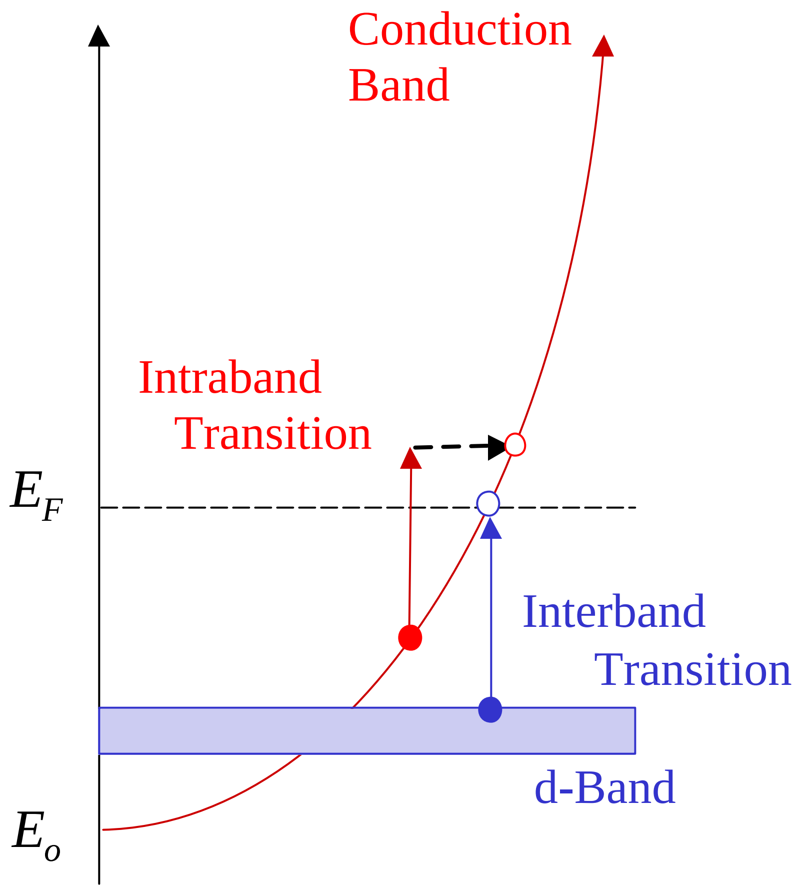
Slower scans of the nonequilibrium period allow for the measurement of electron phonon coupling factor, *G*. A linear relationship was used, $DR/R = aDT_e + bDT_l$.

Electron Phonon Coupling Factor (Au)



Note that the values of the electron-phonon coupling factor determined assuming a linear reflectance model change with the incident fluence.

Absorption Mechanisms in Metals



Intraband Transitions

1) Indirect transitions which require a collision.

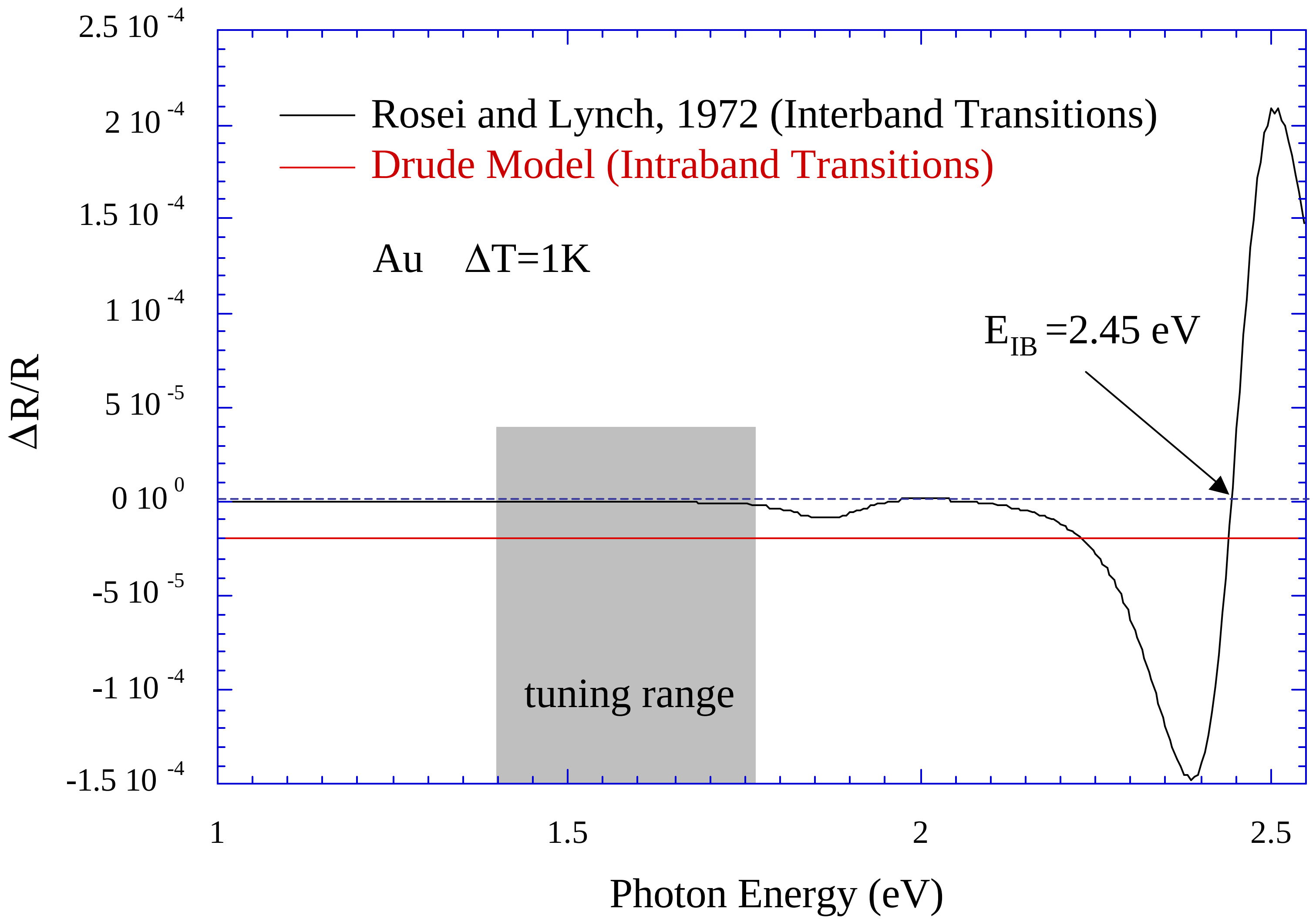
2) Strongly influenced by the electron collisional frequency.

Interband Transitions

1) Direct transitions from the filled d-band to the conduction band.

2) Strongly influenced by changes in both the Fermi distribution and the interband transition energy.

ThermoReflectance Response of Au



In the tuning range of the Ti:Saph laser, the thermoreflectance response of Au results from changes in the intraband transition probabilities.

Intraband Transitions

Drude model for the dielectric function of a nearly free electron metal

$$e(T_e, T_l) = 1 - \frac{w_p^{\;2}}{w\left[w + i w_t (T_e, T_l)\right]}$$

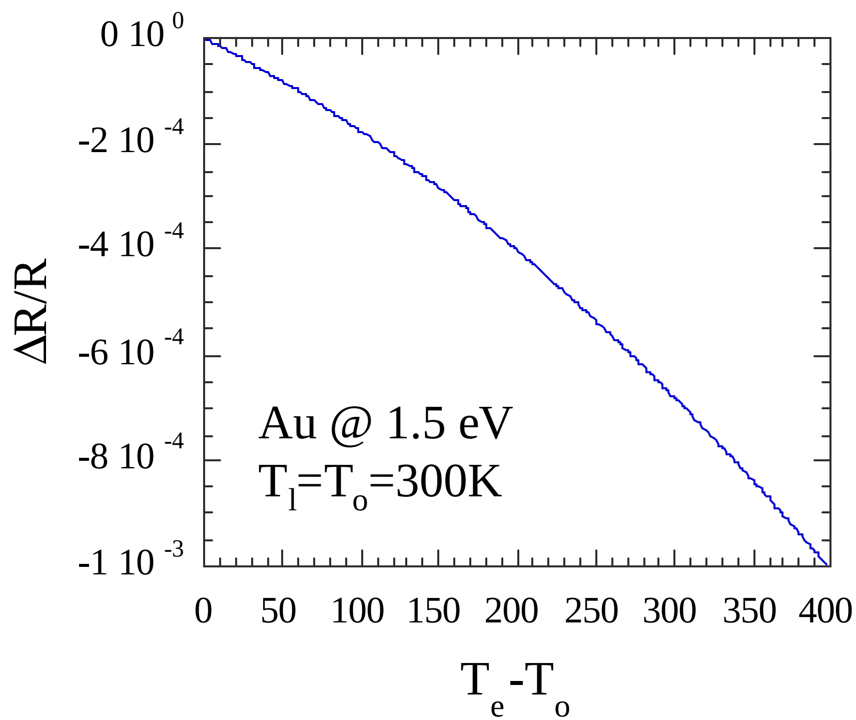
w_p Plasma Frequency

w Incident Photon Frequency

Electron Collisional Frequency:

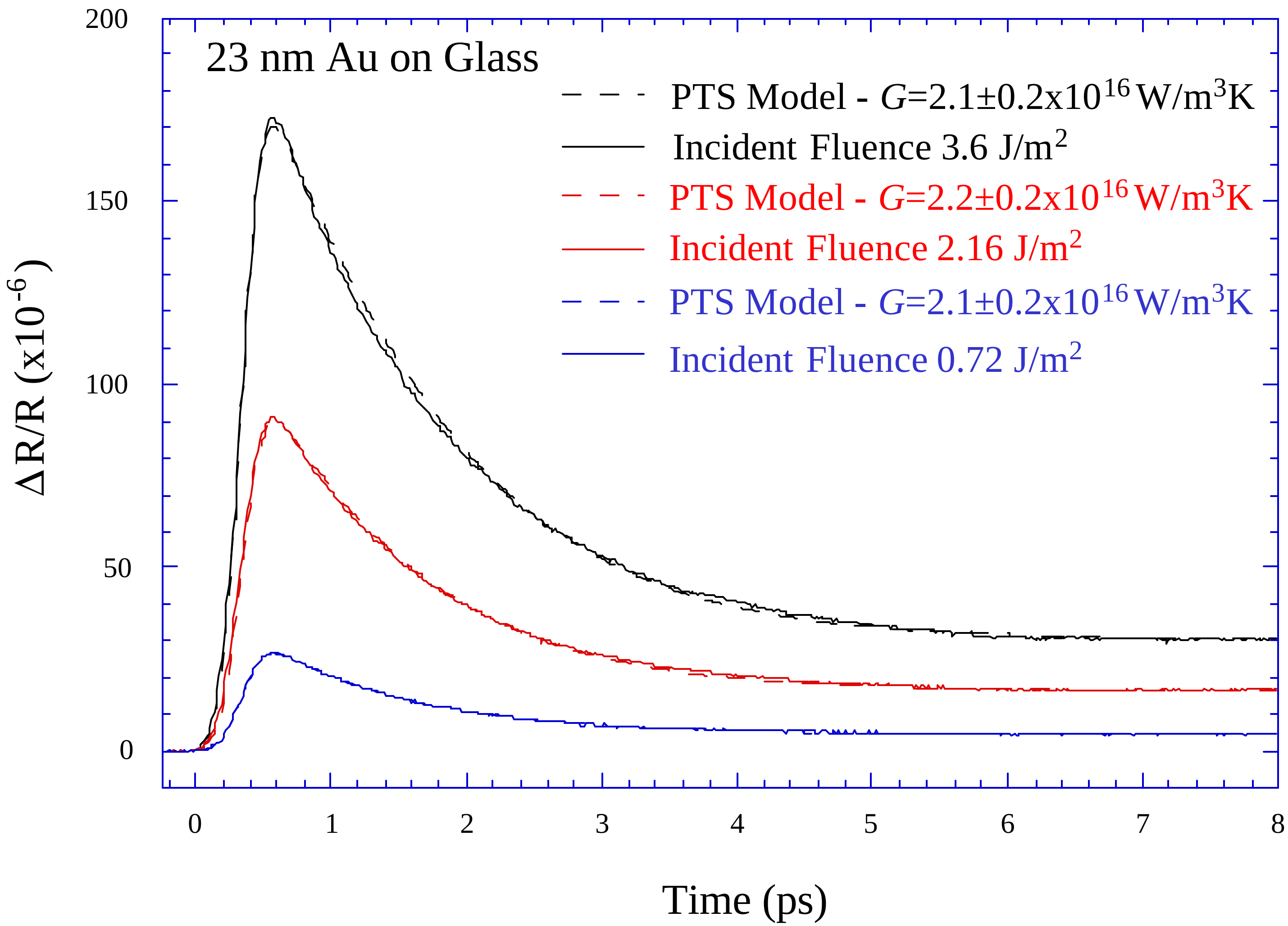
$$w_t (T_e, T_l) \approx \frac{1}{t} = A_{ee} T_e^{\;2} + B_{ep} T_l$$

$$DR \approx 10^{-6} \;\; \frac{1}{\mathrm{K}}$$



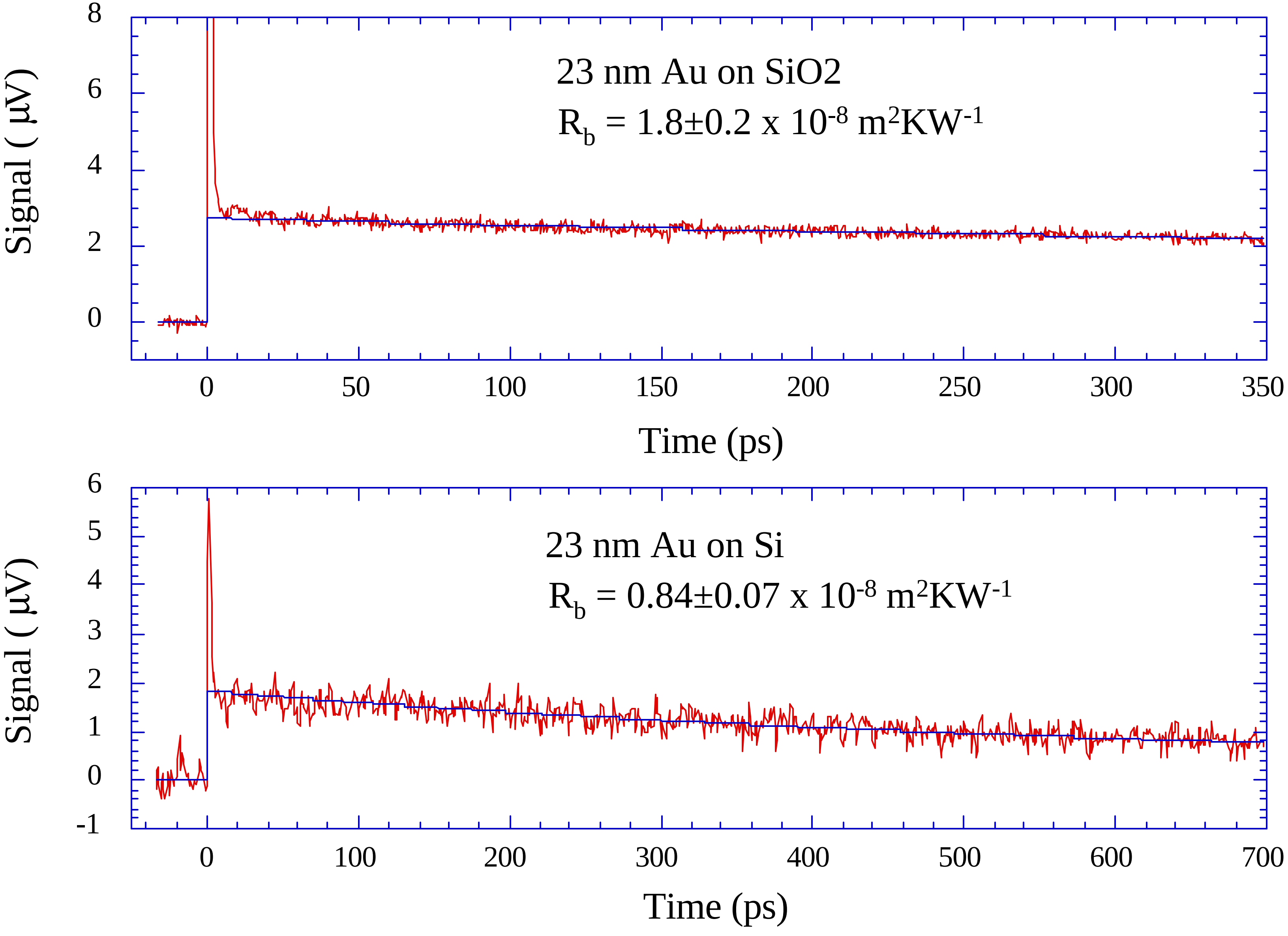
Nonequilibrium Electron Temperature

Intraband Reflectance Model (Au)

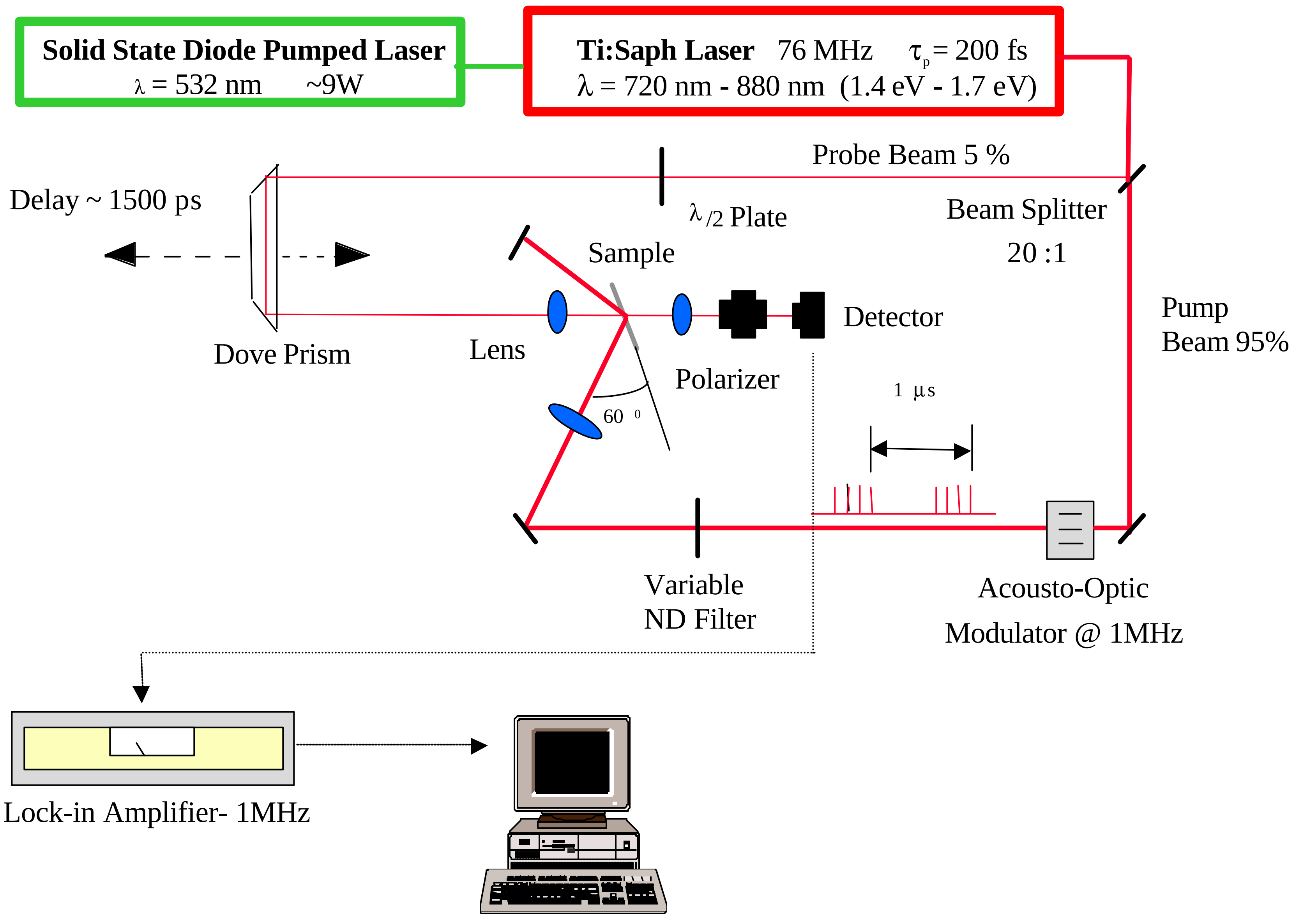


The Drude model was used to relate changes in temperature to reflectance. The electron-electron scattering coefficient was determined to be A_{ee} = 1.4x10^7 1/sK^2.

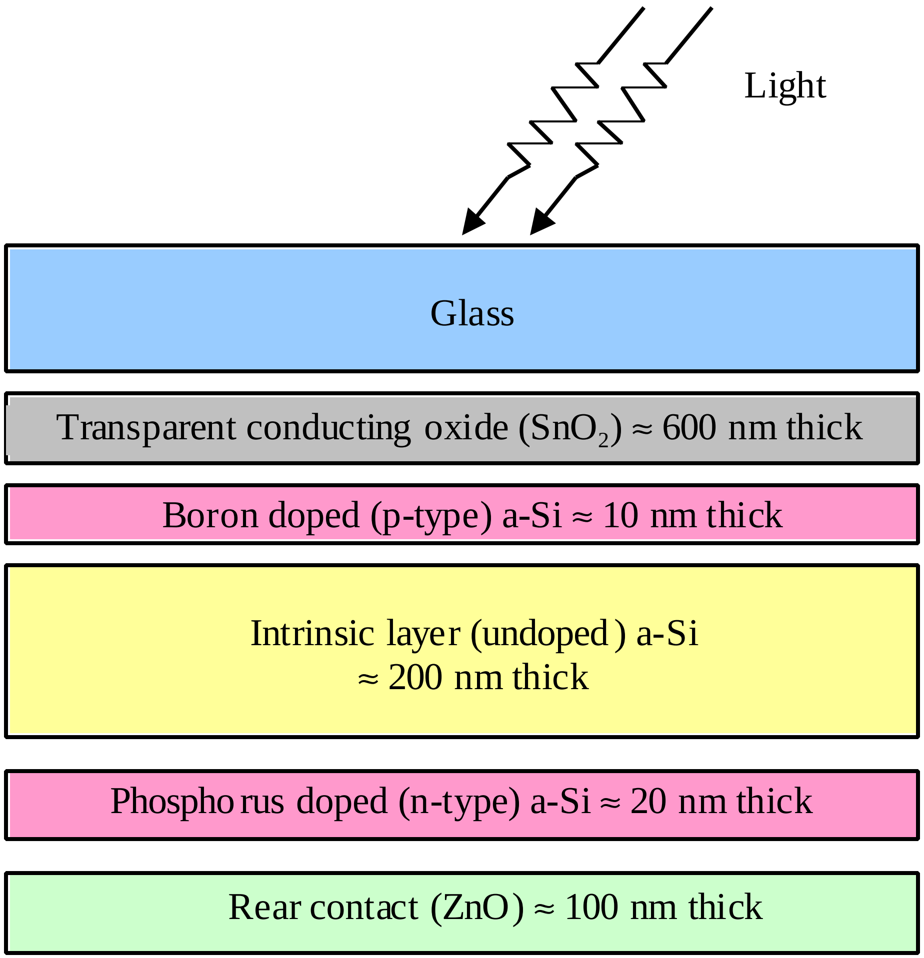
Thermal Boundary Resistance



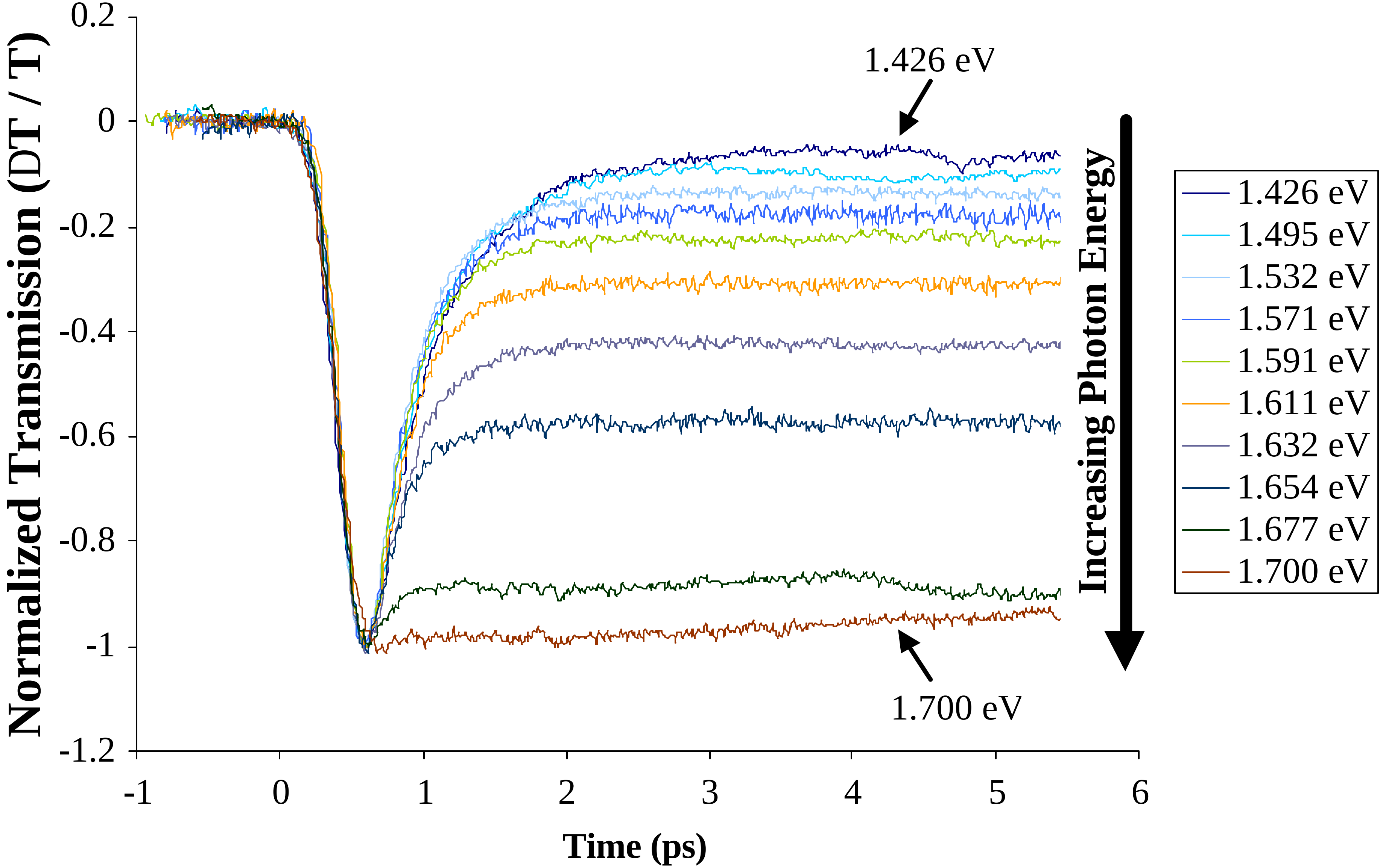
Transient ThermoTransmittance Technique



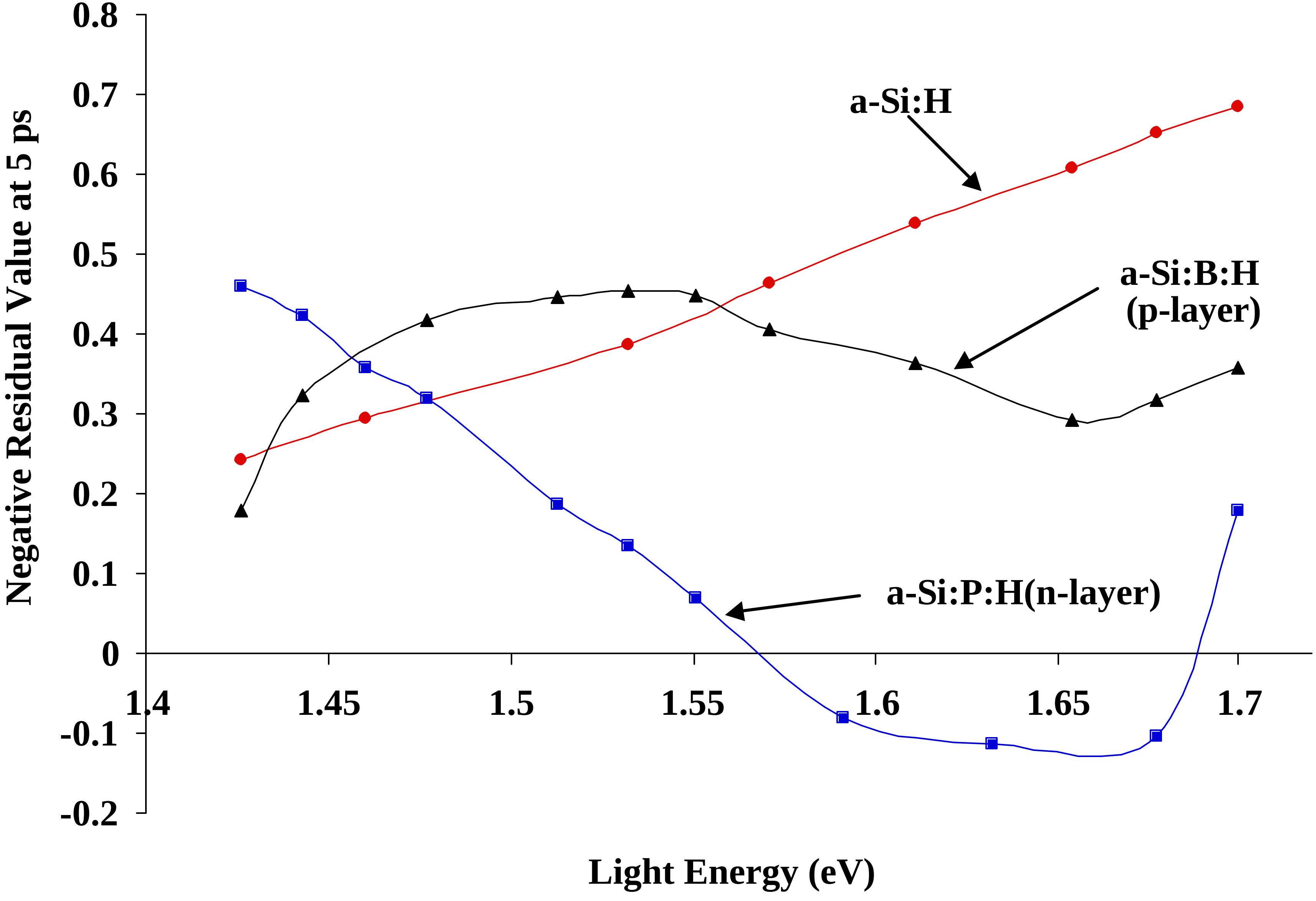
Probing Amorphous Silicon Solar Cells



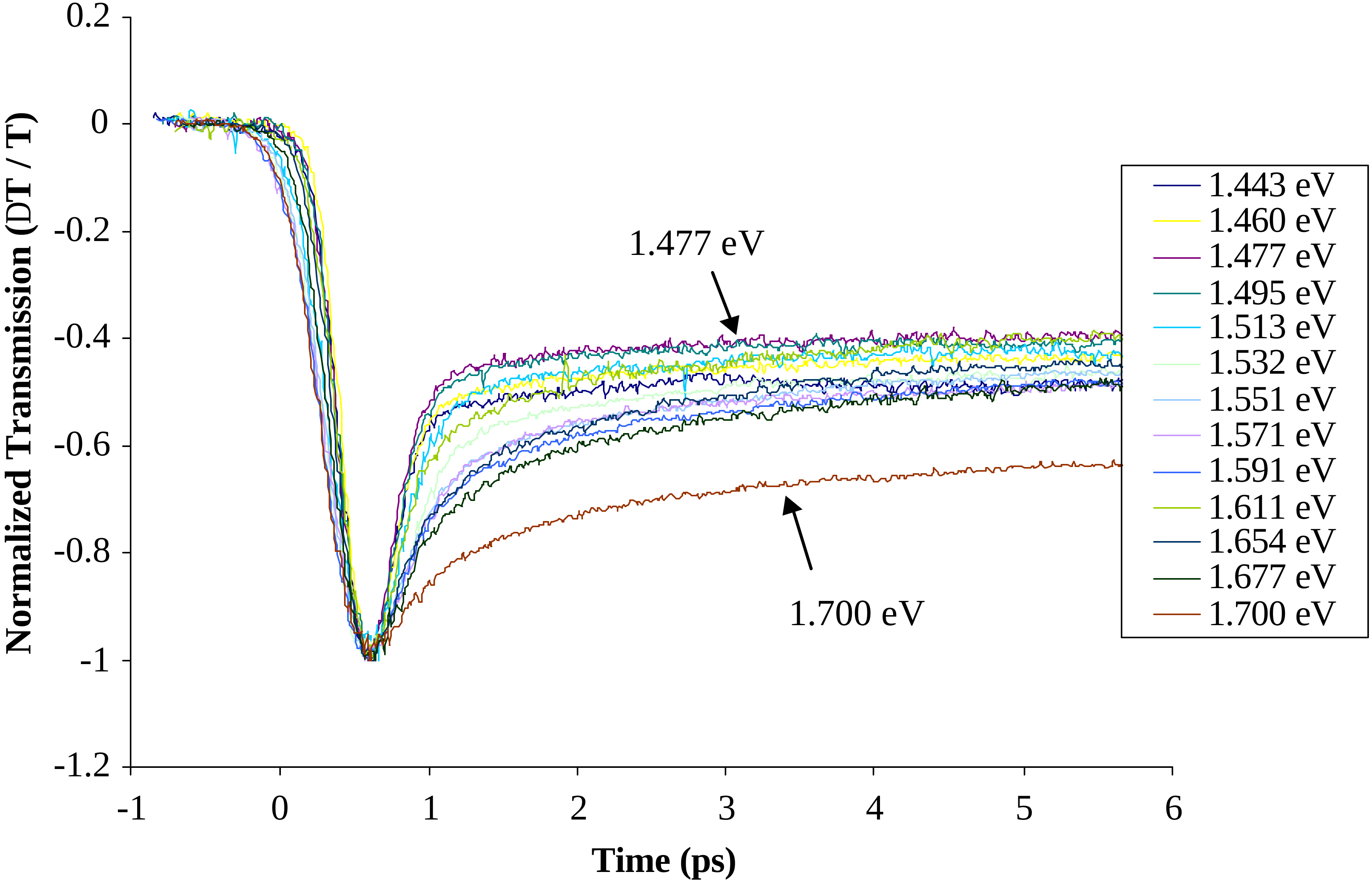
Scans of the 200 nm Intrinsic a-Si Layer



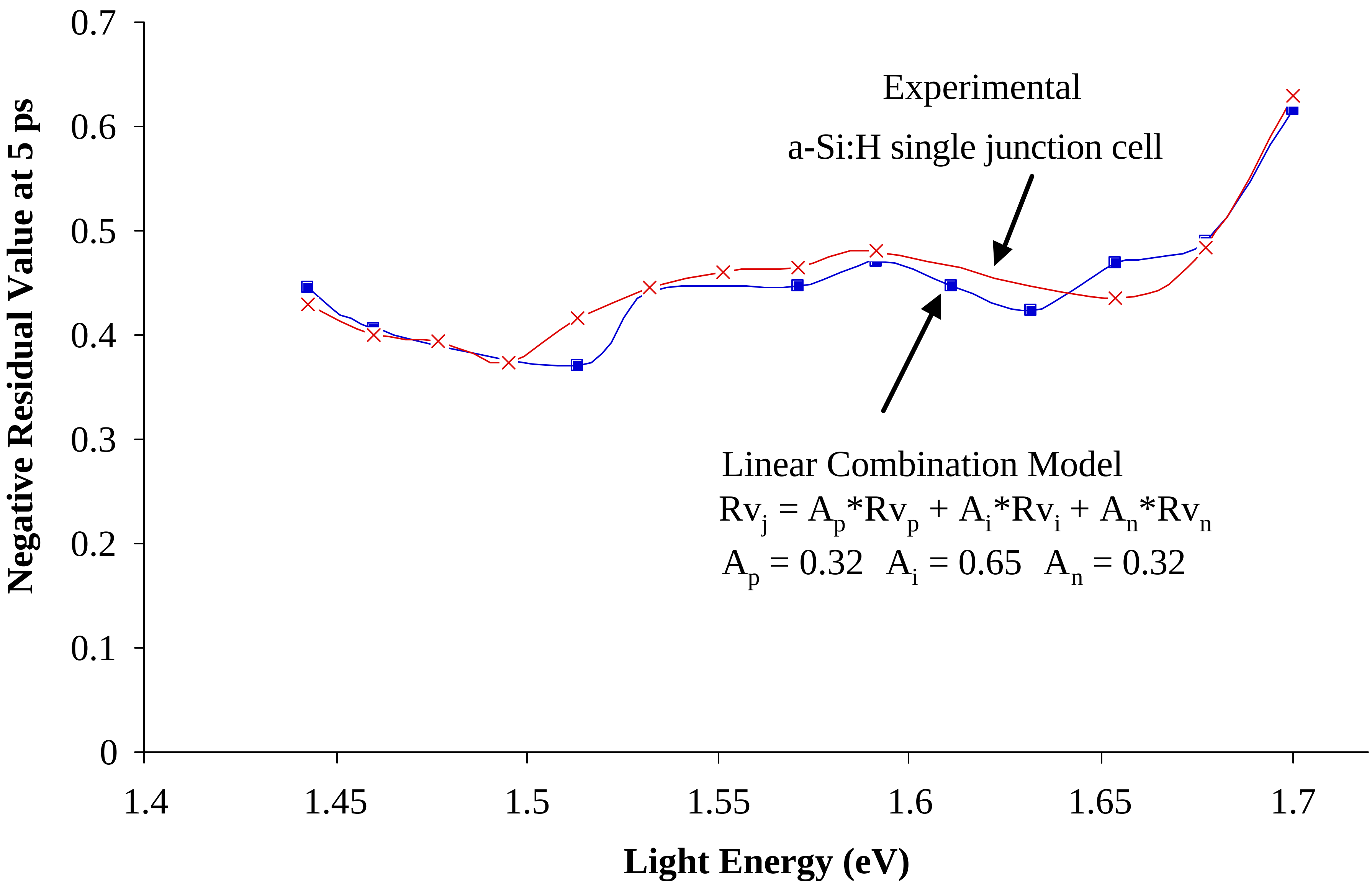
Residual Values for Individual Layers



Scan of a Single Junction a-Si:H Cell



Residual Value for Junction vs. Model



Distinguishable Material Parameters

- **Bandgap**
- **Sample composition (germanium and carbon alloys)**
- **Dopant concentration (phosphorus and boron doping)**
- **Hydrogenation level (defect passivation / crystallinity)**

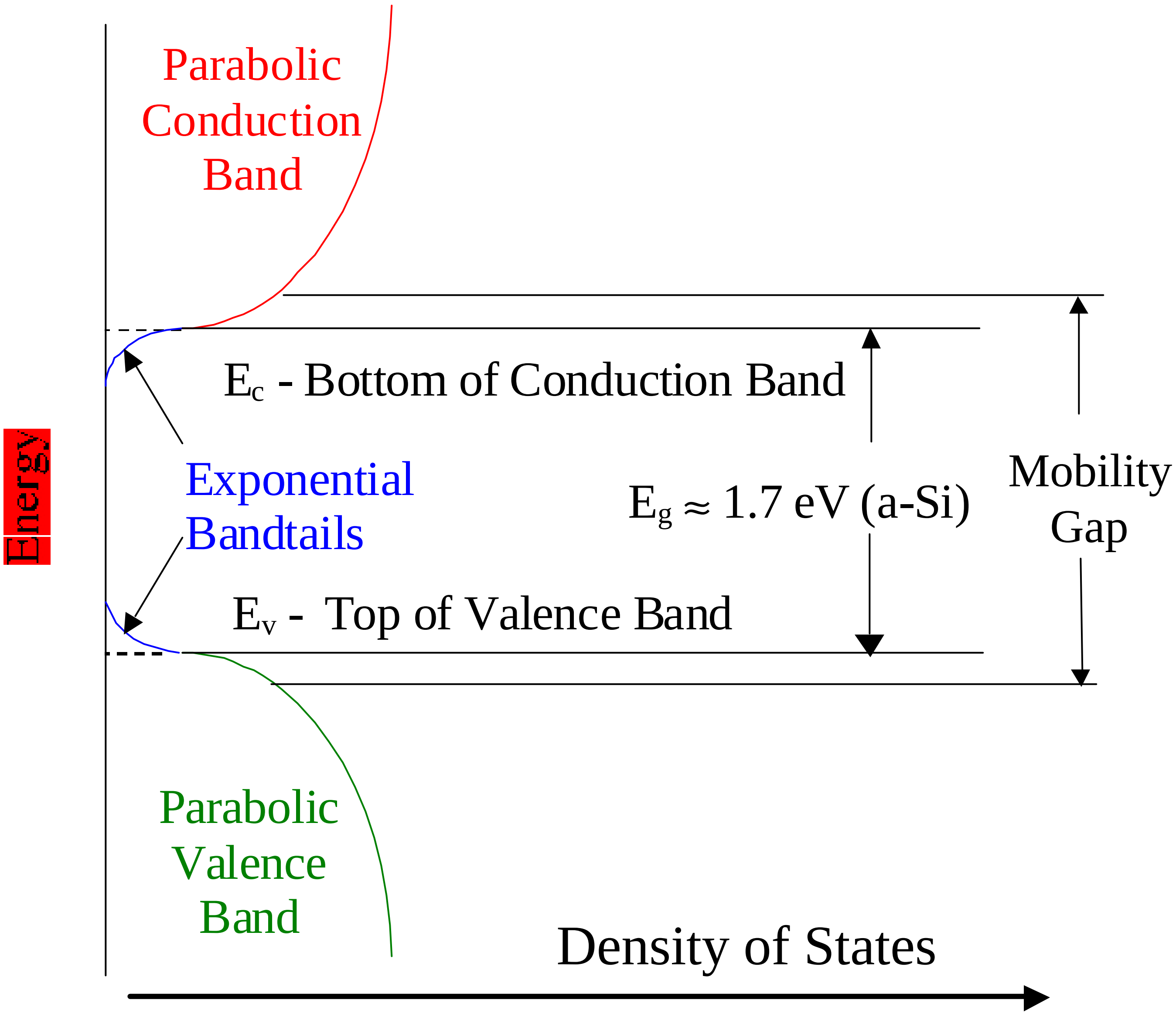
Conclusions

- Presented Femtosecond Transient ThermoReflectance (FTTR) technique measurements of the thermal diffusivity, electron-phonon coupling factor, and thermal boundary resistance of thin metallic films.

- Showed that the thermoreflectance response is two orders of magnitude greater when the incident probe energy is near an interband transition, and that the response is only linear for small changes in temperature.

- Demonstrated that the wavelength dependence of Femtosecond Transient ThermoTransmittance (FTTT) response from an amorphous silicon solar cell can be related to the response of the individual layers.

Simplified Band Structure of a-Si



Model Assumptions

- **Change in transmission is entirely due to change in absorption.**
- **Band structure is parabolic with exponential band-tails.**
- **Absorption before and after spike is “interband” absorption into band-tails. Difference is due to temperature increase.**

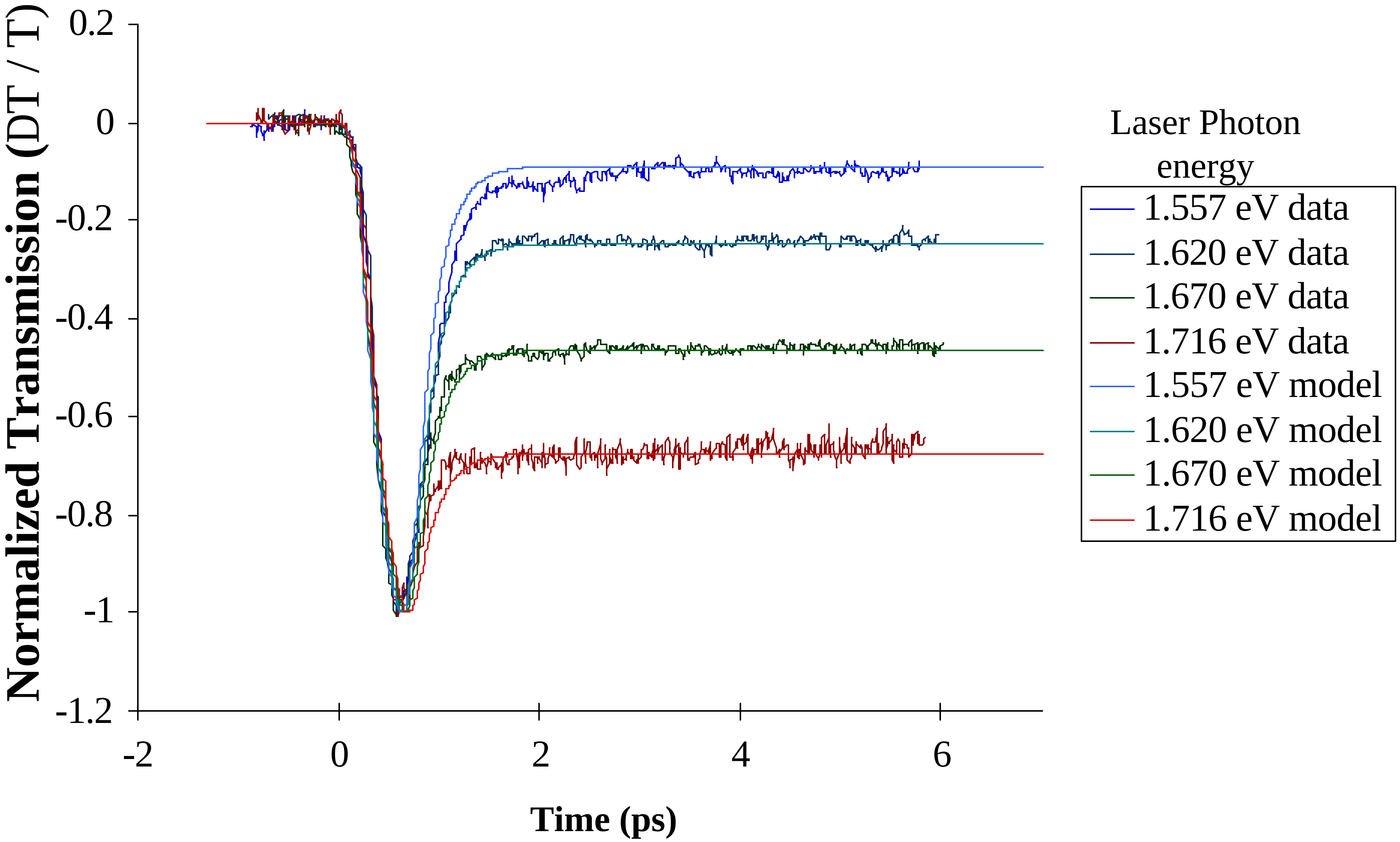
$$N = G_o - G_o \exp(-a x)$$

$$a_T = \alpha_o \exp\{[hn - (E_o - bT)] / 2k_B T\}$$

- **Spike is due to “intraband” absorption of free carriers generated by the pump being excited higher within the conduction band.**

$$a_{fc} = l^2 \frac{q^3}{4p^2 c^3 n^* e_0} \left(\frac{N_n}{m_n^2 m_n} + \frac{N_p}{m_p^2 m_p} \right)$$

Experiment-Model Comparison



Effects of Electron-Electron Scattering on Thermal Conductivity

Electron Thermal Conductivity

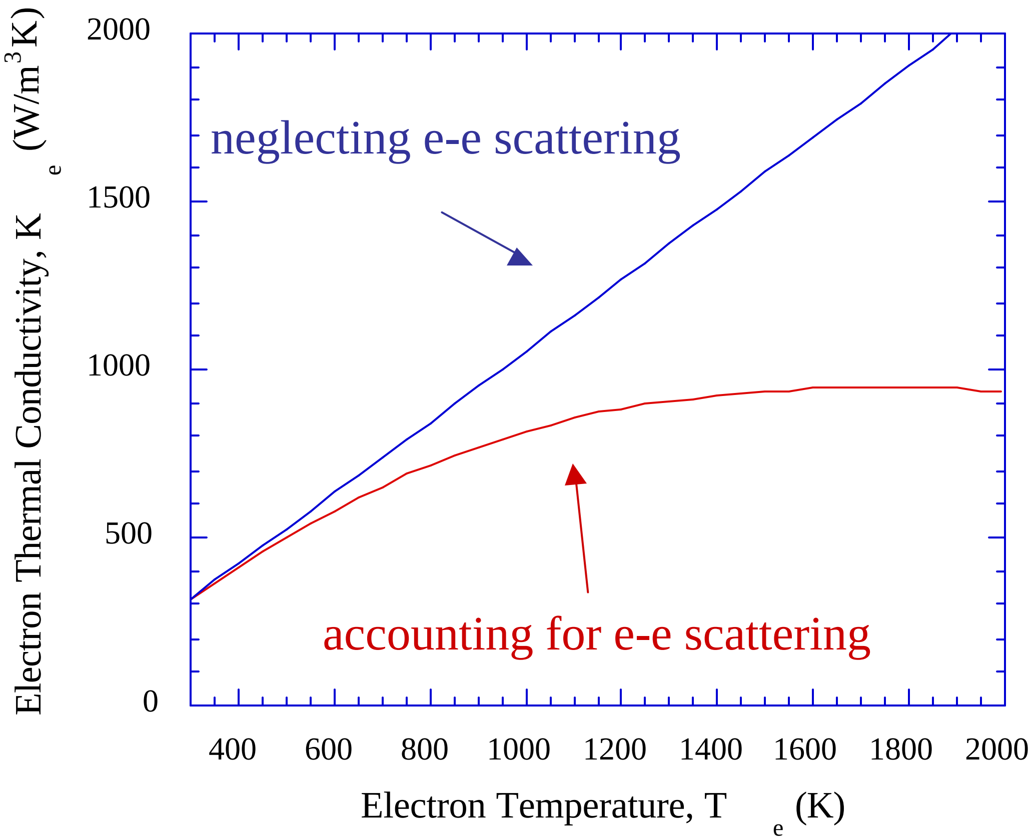
$$K_e \propto \frac{C_e}{w_t}$$

Electron Heat Capacity

$$C_e = gT_e$$

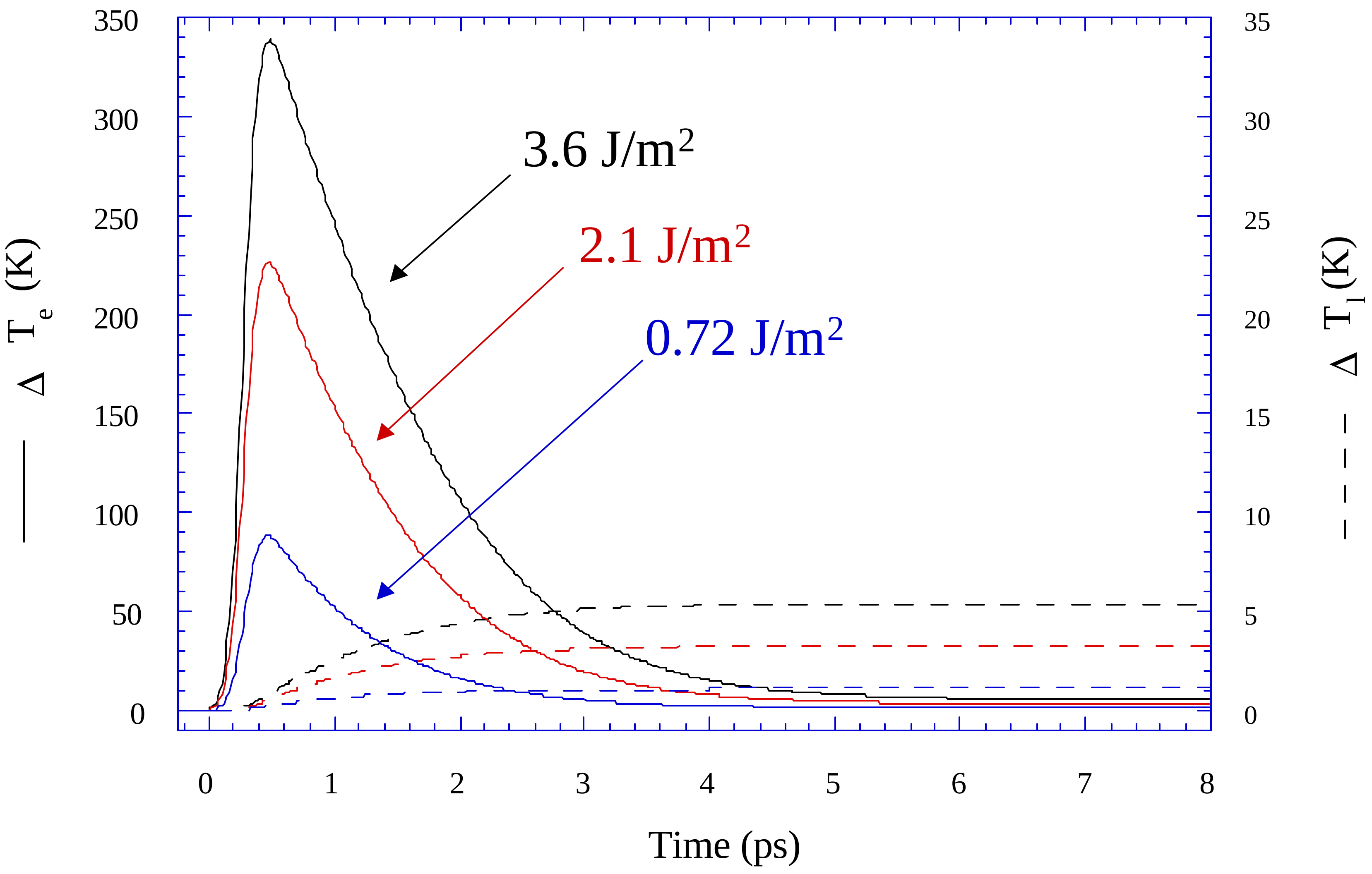
Electron Collisional Frequency

$$w_t = A_{ee}T_e^2 + B_{ep}T_l$$

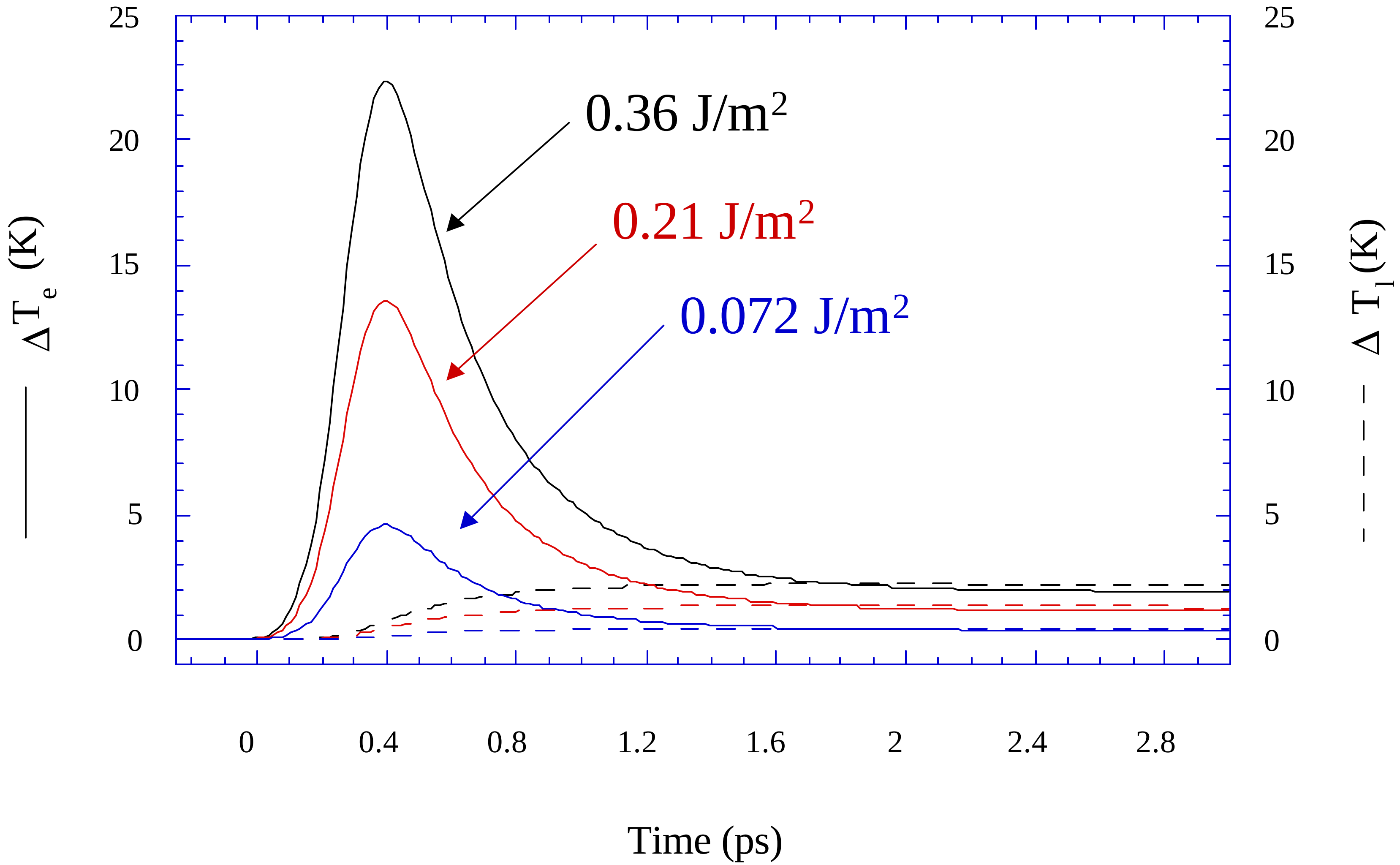


Electron-electron scattering becomes important at higher electron temperatures.

Predicted Temperature Response, Au



Predicted Temperature Response, Pt



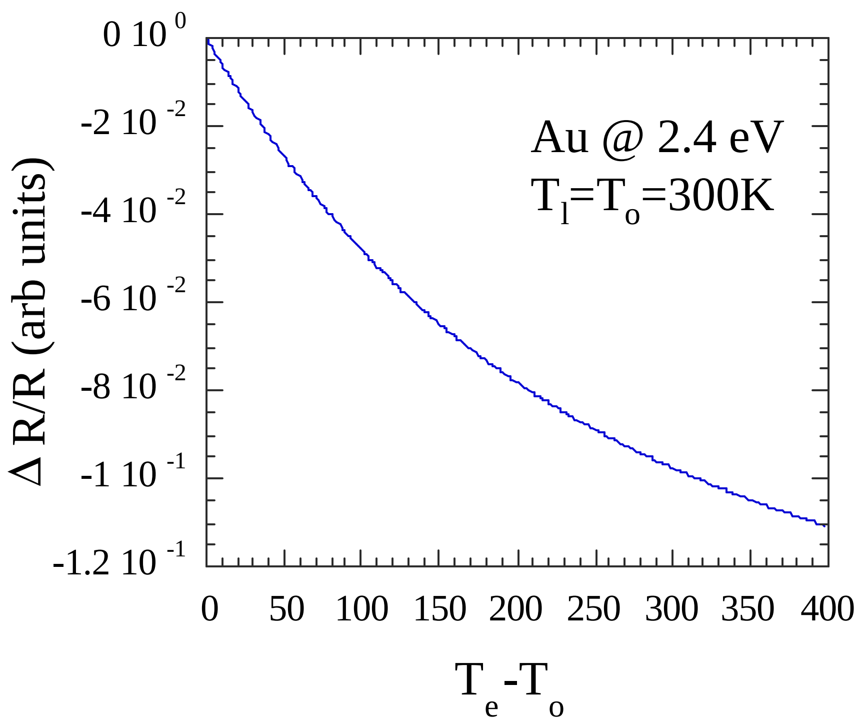
Interband Transitions

Rosei and Lynch, 1972

$$e_2 \propto \frac{(E-E_o)^{1/2}}{E^2}\left(1-\frac{1}{1+e^{\frac{(E-E_1)}{k_B T}}}\right)$$

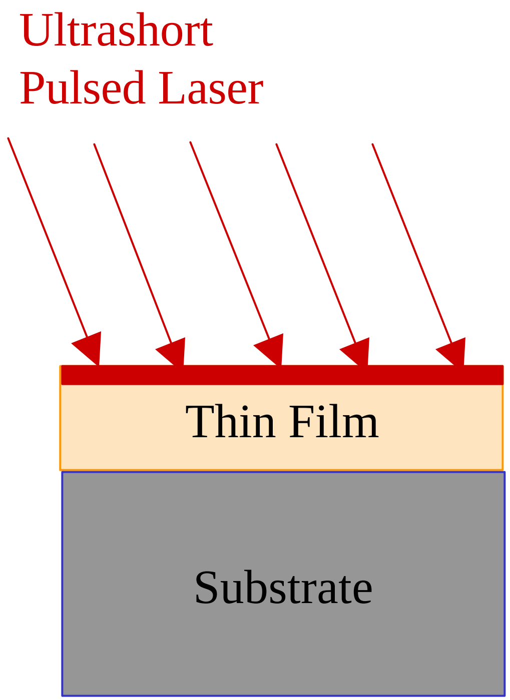
Using the Kramers-Kronig relations this expression can be used to calculate the temperature dependence of the complex dielectric function.

$$DR \approx 10^{-4}\ \frac{1}{\mathrm{K}}$$

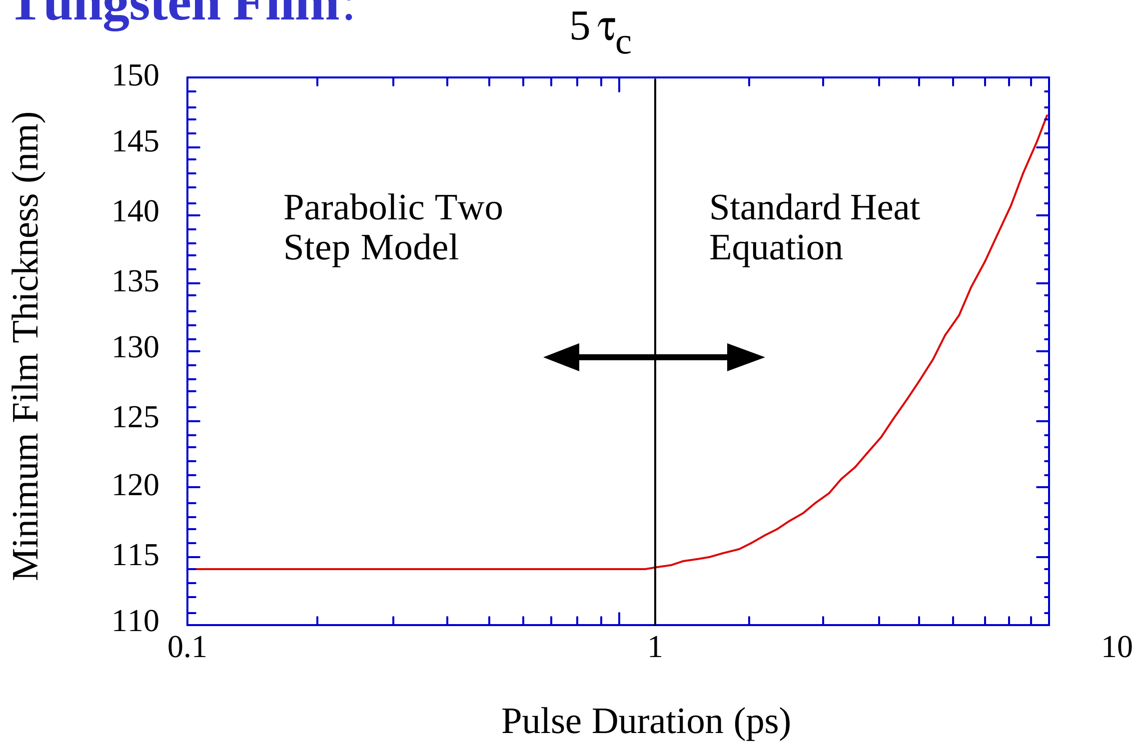


Nonequilibrium Electron Temperature

Transient Thermal Measurement



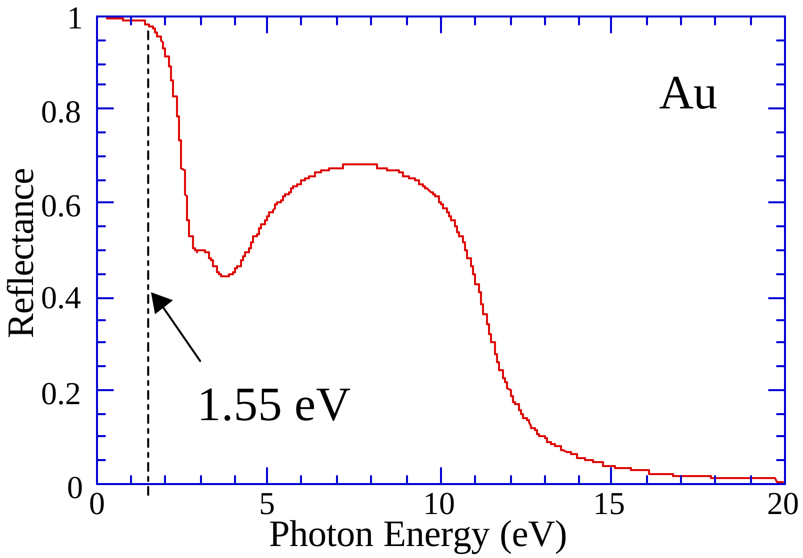
Tungsten Film:



Thermal Penetration Depth:

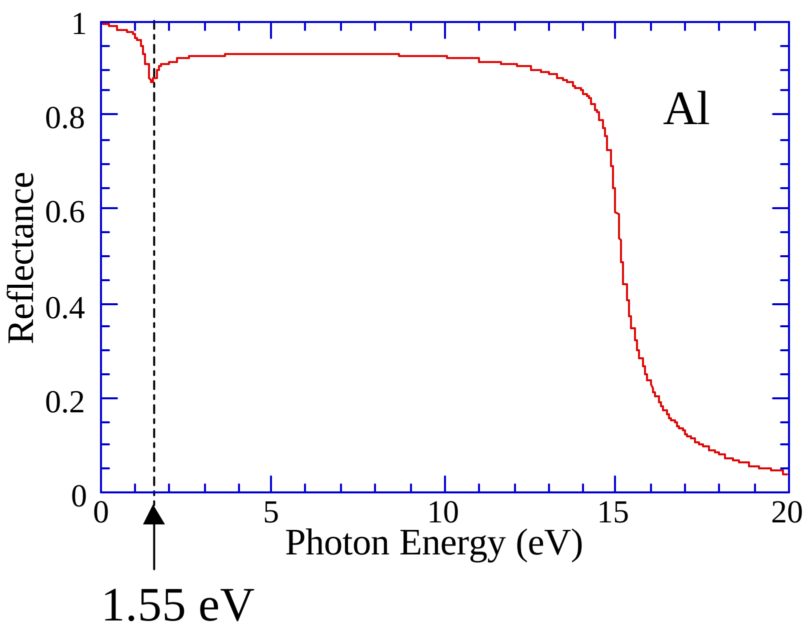
$$d_{thermal} \approx \left(d_{optical} + \sqrt{\frac{k_e t_c}{C_e}} + \sqrt{\frac{k_e t_{pulse}}{C_l}} \right)$$

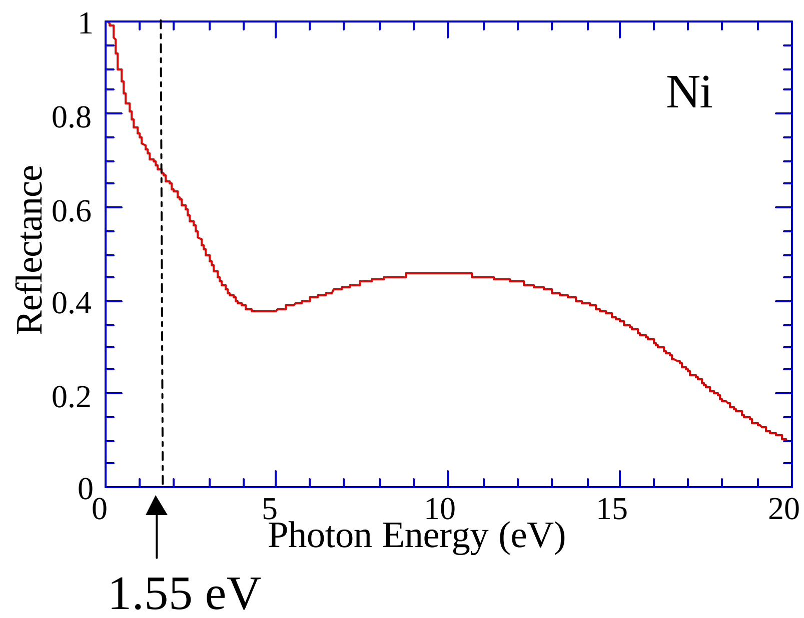
Interband Effects on Reflectivity



If probe photon energy is near an interband transition, then the bound electron contribution can be significant.

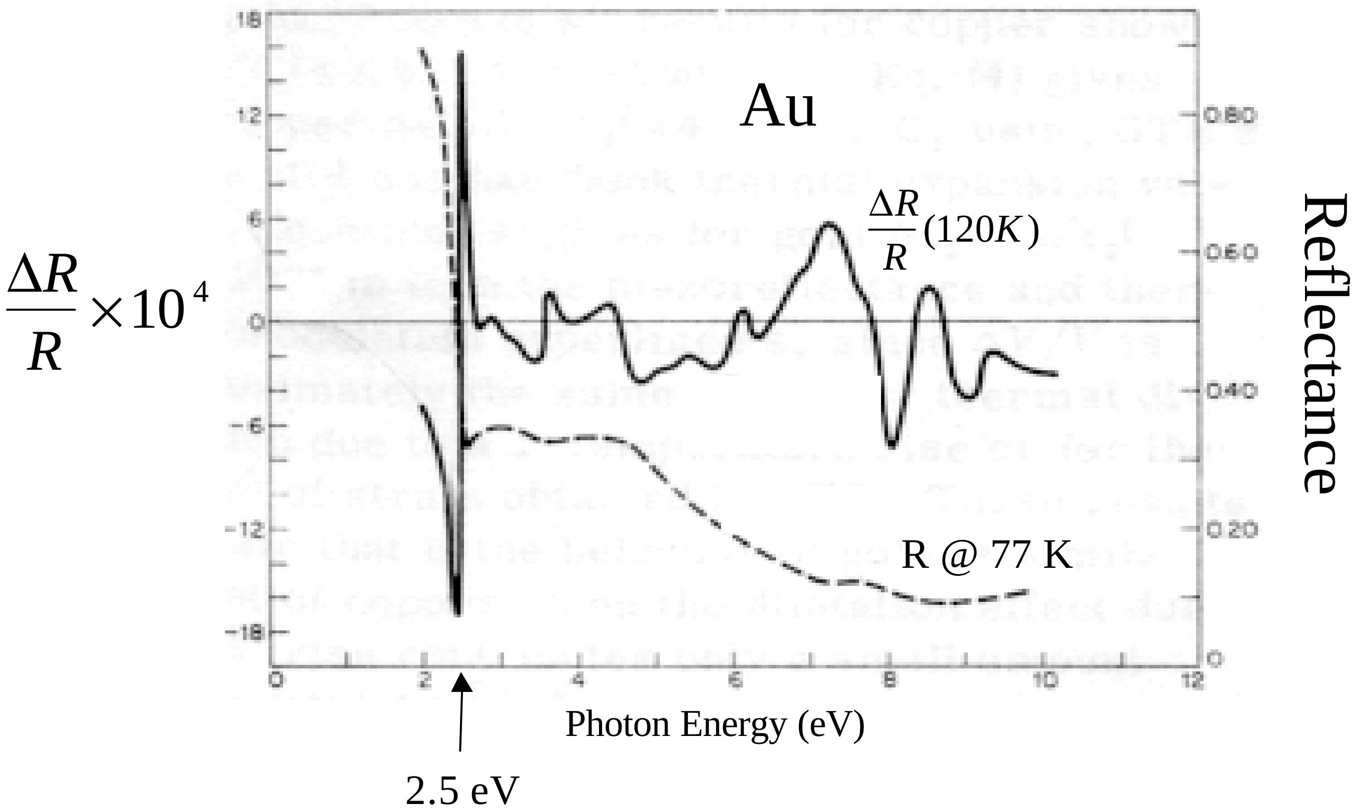
Reflectance spectra calculated from the Drude - Lorentz model with constants from Rakic et al., *Appl. Optics*, **37**, 5271, 1998.





Thermomodulation Critical Points

(Scouler, *Phys. Rev.*, Vol. 18, No. 12, p. 445, 1967)

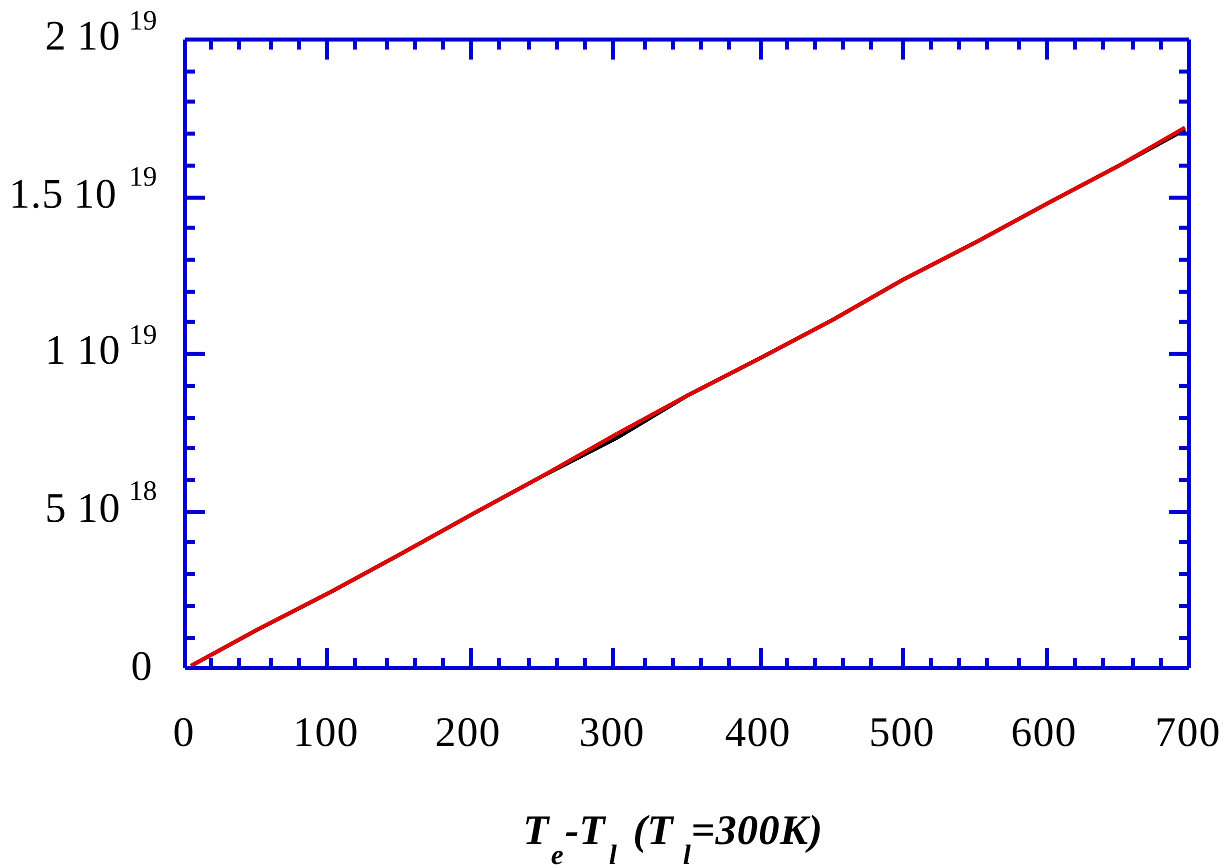


Interband effects cause signal polarity reversal.

Electron Phonon Coupling

Calculated from the electron-phonon collisional equations using the Debye model and assuming that $T_e > T_l > T_{D.}$

$$\left[\frac{\partial U_l}{\partial t}\right]_{ep} Wm^{-3}$$



Ultrashort Pulsed Laser Heating

Parabolic:
heat propagation is diffusive

One Step:
electrons and lattice are in equilibrium

Hyperbolic:
heat propagates at a finite speed

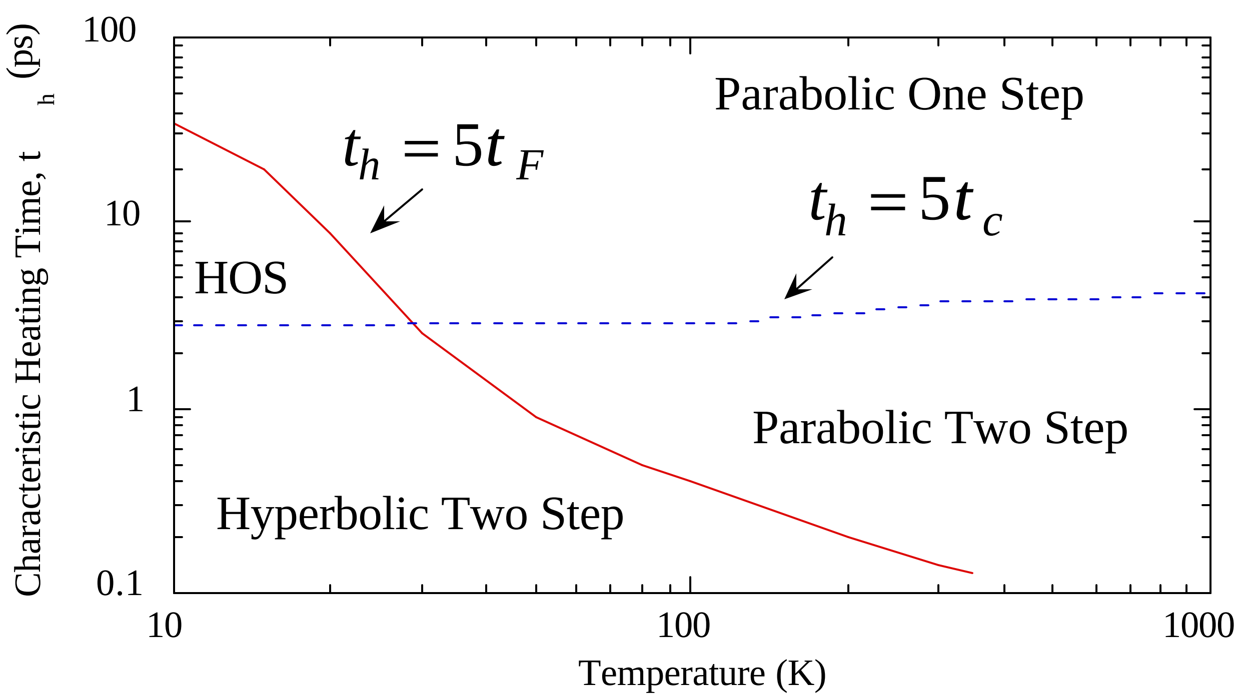
Two Step:
energy is initially absorbed by electrons then coupled into the lattice

Regime Map: (Au)

Thermalization Time:

$$t_c = \frac{C_e C_l}{(C_e + C_l)G}$$

$$\approx \frac{C_e}{G}$$



Nonlinear Thermoreflectance Response